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Radiological Protection in Industries Involving NORM: A (Graded) Methodological Approach to Characterize the Exposure Situations

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Abstract: The interest in radiation protection in industrial sectors involving Naturally Occurring Radioactive Materials (NORM) is increasingly growing. This is due also to the recent implementation of the European Council Directive 59/2013/Euratom which in Italy and in the other European Union Member States extends the field of application to industrial sectors never involved before. This paper reports main results of a research project on radiation protection in industries involving NORM carried out in Italy aimed to provide useful tools for stakeholders to comply new legal obligations. The project activities were mainly focused on different aspects relevant to the NORM involving industries, accounting for the positive list reported in the Italian law. Firstly, the inventory of the industries currently operating in Italy in order to identify the industrial sectors with an important radiological impact on population and workers was updated. Based on this information, a general methodology was elaborated taking into account a graded approach. The first phase consists in the identification and characterization of the most critical exposure scenarios and of the radiological content of NORMs involved in the different phases of the industrial processes. In the second phase calculation methods were developed for dose estimation for workers and members of public. These tools require the use of existing and well tested calculation codes, and the development of a dedicated user-friendly software.

Keywords: NORM; inventory; methodology; exposure scenarios; radiological protection

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1. Introduction

Some radionuclides occur naturally in the environment, since present in the terrestrial crust, and in many materials, used directly (e.g., building materials) or as raw materials for industry [1]. These materials of terrestrial origin are referred to as naturally occurring radioactive material (NORM). Certain industrial activities can increase the potential for human exposure to the NORM radionuclides, sometimes generating residues with radionuclide concentrations above natural background levels. The number of industrial sectors involving NORM is large, for example non uranium extraction activities, coal combustion plants, water treatment plants, zircon and zirconia industry, geothermal energy production, etc. Radionuclides of most concern are elements of the U-238 and Th-232 decay series, and K-40. NORMs can occur in active industries, as well as in former facilities and activities (i.e., legacy sites). If NORMs are not properly managed, they can determine exposures of radiological concern for workers, members of the public, and the environment. From some decades International Bodies and Institutions, like IAEA [2], European Union [3] and ICRP [4] have addressed the "NORM issue" and published many guidelines, documents, recommendations and regulations. As consequence of that international interest, many countries have developed national legislations to ensure proper management of NORM. However, even in some European Union Member States characterized by a well-developed radiation protection regulatory system, there are still challenging issues associated with the management of NORM involving industries. An extensive body of literature relevant to NORM management is available, including documents prepared by IAEA [5–12] and by European Commission [13–15].

In Italy, with the transposition of Council Directive 59/2013/Euratom [3]—also defined as Euratom Basic Safety Standard (EU-BSS)—the regulatory system on radiation protection introduced some novelties regarding exposure in industries involving NORM [16]. The most notable ones are: (i) considering the NORM industries as Planned Exposure Situations (PES); (ii) extension of the field of application to new industrial sectors compared to the list of NORM activities from the previous regulation [17] (iii) the introduction of Exemption and Clearance Levels (EL/CL), in terms of activity concentration and dose, as operational radioprotection tools.

Given the large number of industrial sectors, there is a need of a general methodological approach providing indications about the characterization of exposure scenarios and of protocols to be adapted to the specific industries of interest. Concerning the general methodology, it can support the involved stakeholders to address many aspects of the NORM issue management, such as: identification of the matrices of interest, the choice of the appropriate measurement methods, the sampling techniques, the models to be applied for the dose assessment. In this context, a research project aimed to develop technical-scientific tools useful for the implementation of the Italian legislation on radiation protection was launched involving some Italian institutions. This paper presents deliverables of the project and discusses about further perspectives.

2. Italian Legislative Framework

According to the EU-BSS 2013, the Italian regulation on NORM introduces two types of ELs/CLs: the first is defined in terms of activity concentration and applies to solid matrices, such as raw materials, intermediates and residues. Tables 1 and 2 summarize activity concentration values set as general and specific ELs/CLs, respectively.

In particular, in Table 1 the value of both the clearance and the exemption levels is equal to 1 kBq/kg for the uranium and thorium series if they are in secular equilibrium with their descendants. If not, the value remains the same except for Po-210 and Pb-210, which becomes 5 kBq/kg. The clearance and exemption level for potassium is 10 kBq/kg.

Values given in Table 2 also consider those situations in which segments of the decay chains are not in equilibrium with the parent radionuclides, situation that frequently occurs in NORM residues. A practice involving NORM is exempted from notification, and the descendant obligations, when NORM activity concentrations comply with ELs and CLs reported in Tables 1 and 2.

In case NORMs do not comply with ELs and CLs in terms of activity concentration, the application of the graded approach allows to demonstrate the compliance with ELs and CLs in terms of annual effective dose for workers and members of the public (see Table 3). The dose criterion for workers is the same established in EU-BSS (1 mSv per year), while for members of the public, Italy, along with few other European countries [18], has adopted a more conservative dose criterion of 0.3 mSv per year. This value takes into account all pathways of exposure, including airborne or liquid effluents and contribution to dose from the disposal or recycling of solid residues.

Table 1. General Exemption and Clearance Levels (ELs and CLs).

Natural Radionuclides		ELs and CLs
Radionuclides from ^{238}U and ^{232}Th series in secular equilibrium	all radionuclides	1 kBq/kg
^{210}Pb and ^{210}Po (when ^{238}U and ^{232}Th series are not in secular equilibrium)		5 kBq/kg
^{40}K		10 kBq/kg

Table 2. Specific ELs and CLs for particular materials or residue destination.

Specific Situations	Radionuclides	ELs	CLs
Oil sludge	U-nat, ^{230}Th, ^{232}Th, ^{210}Pb, ^{210}Po	100 kBq/kg	100 kBq/kg
	^{228}Ra	10 kBq/kg	10 kBq/kg
	For all radionuclides of the ^{238}U and ^{232}Th series	5 kBq/kg	5 kBq/kg
	^{40}K	50 kBq/kg	50 kBq/kg
Disposal in landfill or reuse for road construction	^{238}U and ^{232}Th series		0.5 kBq/kg
	^{210}Pb and ^{210}Po		2.5 kBq/kg
	^{40}K		5 kBq/kg
Incineration			Dose assessment for members of the public
Disposal of residues or effluents with potential impact on drinking water sources			Dose assessment for members of the public

Table 3. ELs and CLs in terms of annual effective dose.

Category	Dose Value
Workers	1 mSv/year
Members of the public	0.3 mSv/year
	0.1 mSv/year in case of potential impact on drinking water sources

Analogously to the case of activity concentrations of solids below ELs from Tables 1 and 2, a practice involving NORM is exempted from notification, and the descendant obligations, when NORM activity concentrations comply with ELs and CLs reported in Table 3.

For residues disposed in landfill or used for road construction, specific activity concentration values have been set at 50% of the general CLs (see Table 2), since the CLs of 1 kBq/kg does not always comply with the dose criterion of 0.3 mSv/y for members of the public.

3. Material and Methods

3.1. Overview and Inventory of NORM Sites and Exposure Scenarios in Italy

International Bodies, like UNSCEAR [19] encourage the construction and continuous updating of national inventories of NORM involving industries. In 2017, under the auspices of the IAEA ENVIRONET network (Network on Environmental Management and Remediation), the NORM project called "Promoting Good Practices and Providing Knowledge Transfer Applicable to the Management of NORM Residues and Wastes" started. A task group has been set aimed at drafting a "Guidance on how to establish a NORM Inventory", to support IAEA Member States in dealing with the challenges related to the radiation protection management of NORM involving industrial sectors [20]. It is worth noting that in the last two decades outcomes of some national inventories were published [21–25]. Indeed, this activity is a useful tool to gather information to assess the radiological impact of NORM involving industries on workers, the public and the environment. Moreover, the national inventory allows to identify still active industrial sectors and relevant secondary processes. The European Project RadoNORM, involving 57 institutions from 22 countries, includes also an activity providing a comprehensive overview of existing NORM sites and their characteristics at the European level and to evaluate the most relevant NORM exposure scenarios and scientific and regulatory questions related to them [26]. As regards the Italian inventory, information about the number of operating NORM involving industries in Italy, their characteristics and distribution at regional and national level, and basic information about the radioactivity content of raw materials and residues was collected.

3.2. The Approach Adopted to Elaborate a General Methodology for the Characterization of the Exposure Sources for Workers and Population from NORM Involving Industries

For practices, the EU-BSS 2013 [3] requires that "the application of a graded approach to regulatory control, which should be commensurate with the magnitude and likelihood of exposures resulting from the practices, and commensurate with the impact that regulatory control may have in reducing such exposures or improving the safety of installations". Later on, the International Basic Safety Standards, published in 2014 by IAEA [2], indicates that "The application of the requirements (. . .) shall be in accordance with the graded approach and shall also conform to any requirements specified by the regulatory body". The concept of graded approach is defined as "(. . .) a process or method in which the stringency of the control measures and conditions to be applied is commensurate, to the extent practicable, with the likelihood and possible consequences of, and the level of risk associated with, a loss of control." The common position of EU-BSS 2013 and the International BSS origin from the ICRP Recommendations 103 of 2007 [27] which reports that "a graded burden of obligation must be foreseen according to the amenability of a particular source or exposure situation to regulatory controls, and the level of exposure/risk associated with that source or situation."

In order to introduce the graded approach in the regulatory control of Italian NORM involving practices, a methodology was developed to characterize the exposure scenarios. This methodology can help stakeholders to assess and manage the NORM involving activities of radiological impact. It is worth noting that stakeholders can be not only the employers of NORM involving industries, but also radiation protection experts, technical figures involved in the control phases, and other possible actors.

Once identified the exposure scenarios, the methodology allows answer some main questions, such as:

1. What are the NORM matrices of radiological concern to be analysed in laboratory and the radionuclide activity concentration to be measured?
2. What are the most suitable analysis methods to be used?
3. Are simplified methods available to assess the exposure of workers and members of the public?

4. Results and Discussion

4.1. Inventory of Italian Industries Involving NORM

A national inventory of industries involving NORM has been built, considering the list of industrial sectors given in the Italian regulation [16] shown in Table 4. It updates and increases data collected in 2014 [28]. From preliminary results (Table 5), a general decrease of number of NORM involving plants emerges. Moreover, some NORM involving sectors (e.g., tile production) are present in the inventory for the first time and for other industries, such as Oil&Gas sector, information is more detailed.

In order to complete information about the radiological impact of Italian NORM involving industries, Table 6 reports mean values of radionuclides activity concentrations in some raw materials and residues, taken from scientific literature and reports of Italian national institutions.

An important result of the survey is that in Italy there are currently no active industries in the following sectors:

- Extraction of rare earths from monazite;
- Extraction of tin, lead and copper;
- Extraction of iron-niobium from pyrochlore;
- Niobite/tantalite processing;
- Production of thorium compounds and manufacture of thorium-containing products.

Table 4. List of NORM involving industrial sectors and type/class of practices included in the Italian legislation.

Industrial Sector	Class of Practice or Critical Exposure Scenarios
Coal-fired power plants	Maintenance of boilers
Mining of ores other than uranium ore	Extraction of granitoids, such as granites, orthogneiss, tuff, pozzolana, basalt, porphyry and lava
Zircon and zirconium industry	Processing of zircon sands Production of refractories, ceramics and tiles Production of zirconium oxide and metallic zirconium
Mineral processing and primary iron production	Extraction of rare earths from monazite Extraction of tin, lead and copper Processing of iron/niobium from pyrochlore ore Extraction of aluminum from bauxite Processing of iron/tantalum Use of potassium chloride as additive in metals extraction by fusion
TiO_2 pigment production	Management and maintenance of titanium dioxide production plants
Processing of phosphate and potassium minerals	Thermal phosphorus production Phosphoric acid production Production and wholesale of phosphate and potassium fertilisers Production and wholesale of potassium chloride
Cement production	Maintenance of clinker ovens
Production of thorium compounds and manufacture of thorium-containing products	Production of thorium compounds and manufacture, management and conservation of thorium-containing products, in particular welding electrodes with thorium, optical components with thorium and nets for gas lamps
Geothermal energy production	Maintenance of high or medium-enthalpy geothermal energy systems
Oil and gas production	Oil extraction and refining, gas extraction, in particular for the presence of muds and scales in pipes and oil containers
Industries equipped with groundwater filtration facilities	Management and maintenance of facilities
Cutting and sandblasting processing	Plants using abrasive sands or minerals

Table 5. Results of the 2022 inventory about industries involving NORM in Italy compared with the survey carried out in 2014.

Industrial Sector	**N. of Plants**	
	2014	**2022**
Cement production:		Total: 54
Integral cycle	Total: 81	32
Grinding		22
Geothermal energy production:		
high and medium enthalpy	34	34
Zircon sands industry:		
Tiles prod	82	131
Refractories prod	37	31
Sanitary ware prod		30
Ceramic glazes and dyes prod		15
Coal-fired power plants	13	6
Titanium dioxide production	1	1
Steel production:		
Integral cycle	2	1
Electric furnace	40	37
Oil & gas production:		
Oil production plants	1642 wells	1581 wells (25 plants)
Gas production plants		193
Refinery		10
Aluminum production	1	1
Processing of phosphate and potassic ores		
Fertilizer production		22

Table 6. Radiological characterization of some NORM raw materials and residues.

Industrial Sector	**Raw Materials**	**Residues**	**Ref.**
	Activity Conc. (Bq/g)		
Cement production: - *Integral cycle*		Scales ^{210}Pb: 0.05–0.14; ^{210}Po: 0.05–0.3	[29]
Geothermal energy production: - *High enthalpy* - *Medium enthalpy*		Tower sludge, Scales deposits, Exhausted sand, Sand blasting dust, Adsorbent, Filtering material All nuclides < 1, but some scales have ^{210}Po and ^{210}Pb > 1	[29]
Zircon sands industry: - *Tiles prod.* - *Refractories products* - *Sanitary ware products* - *Ceramic glazes and dyes products*	Zircon sand ^{238}U: 1830–3600 ^{232}Th: 370–520	Tile hydrated lime ^{210}Po: 4.7–46 Refractory Fusion furnace dust: ^{210}Pb: 21, ^{210}Po: 35 Grinding dust: ^{238}U: 1.2, ^{210}Po: 1.3 Sludge ^{238}U: 1.7, ^{210}Pb and ^{210}Po: 1.2	[30]
Coal-fired power plants		Fly ash ^{238}U: 0.08, ^{232}Th: 0.08, ^{210}Po: 0.09, ^{210}Po: 0.08 Bottom ash ^{238}U: 0.14, ^{232}Th: 0.14, ^{210}Po: 0.03, ^{210}Po: 0.05	[29]
Titanium dioxide production		Filtering clothes: ^{232}Th series: 1.5	

Table 6. *Cont.*

Industrial Sector	Raw Materials	Residues	Ref.
Steel production: - *Integral cycle* - *Electric furnace*		Blast furnace dust ^{210}Pb and ^{210}Po: 0.5–1.6 Sintering dust ^{210}Pb: 47.3, ^{210}Po: 42.9	[30,31]
Oil & gas production: - *Oil production plants* - *Gas production plants* - *Refinery*		Sludge ^{226}Ra: 0.6–2 Scales ^{226}Ra: 0.12–3	[30]
Aluminum production		Red mud ^{238}U: 0.10, ^{232}Th: 0.12, ^{226}Ra:0.09	[30]
Processing of phosphate and potassic ores - *Fertilizer production*	Phosphorite ^{238}U: 1.8–3.6, ^{226}Ra:1.1–1.6	Phosphogypsum ^{226}Ra:1.3–4.0	[30]

4.2. Description of General Methodology

The proposed general methodology consists of two Phases, each of one divided into 4 consecutive steps, which, applying the 'graded approach', allow to identify the most critical exposure scenarios and to verify the level of exposure of workers and members of the public, to be compared with the relevant ELs. The Phase 1 is finalized to verify the compliance of solid matrix activity concentrations with ELs in terms of kBq/kg (see Tables 1 and 2). If the exemption is not demonstrated in the Phase 1, the compliance with the ELs in terms of effective dose has to be assessed in the Phase 2. Details of these procedures are described in the following sections.

4.2.1. Phase 1

The four steps of Phase 1 are shown in Figure 1. The first step is aimed to identify the "practice": where the regulation indicates as a practice the entire industrial process, an in-depth study of the industrial process allows identifying the most critical exposure situations.

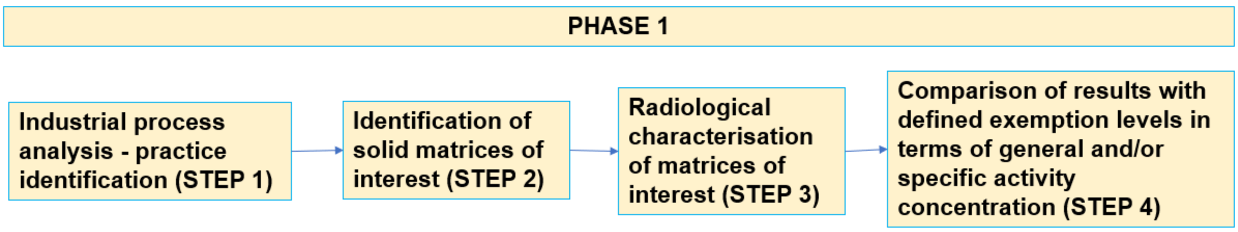

Figure 1. Scheme of Phase 1 of the general methodology to identify the most critical exposure scenarios in NORM involving industrial process.

In step 2, the identification of NORM matrices of interest is carried out: the Italian legislation defines "matrix" as "any substance or material that may be contaminated with radioactive materials; this definition includes environmental matrices (including air, water and soil) and food." Accordingly, raw materials, residues, liquid and gaseous effluents, and final products may be matrices to be considered in the industrial sectors involving NORM. Figure 2 shows a summary of the main categories of matrices of interest. Step 2 is focused on identifying the solid matrices involved in the industrial process and the relevant radionuclide activity concentrations to be measured.

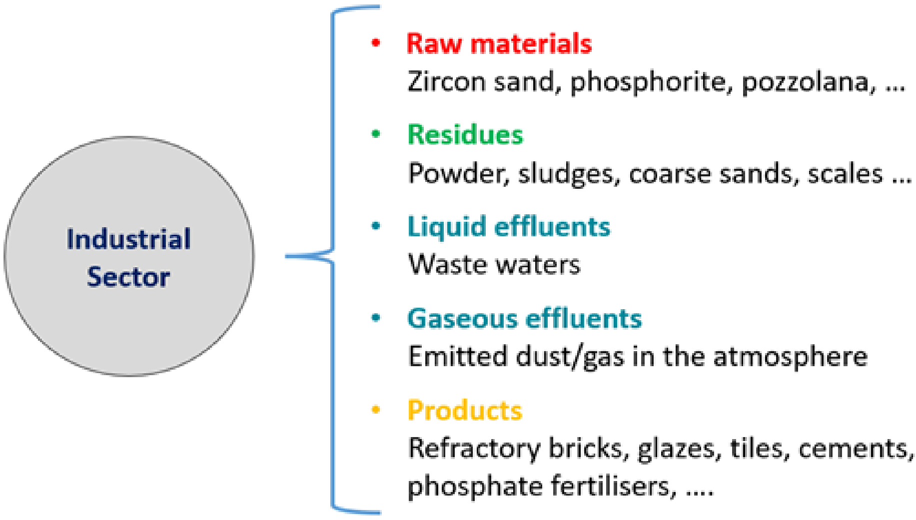

Figure 2. Scheme of NORM matrices considered in the procedure for a certain industrial sector.

Once identified the matrix of interest, measurements of the relevant radionuclide activity concentration are carried out (step 3). In case of a raw material of natural origin, the secular equilibrium among radionuclides of the ^{238}U and ^{232}Th series can be expected. Otherwise, if the raw material has undergone previous industrial processes, the secular equilibrium of radionuclides is not guaranteed. Indeed, it is worthy to note that the chemical/physical characteristics of the industrial processes can determine the disequilibrium of decay chains and influence the activity concentration of single radionuclides (step 3).

Finally, in the step 4, the results of measurements are compared with relevant ELs, shown in Tables 1 and 2.

Table 7 represents the general scheme to support the application of Phase 1; the table items must be specified case by case accounting for the particular industrial sector under analysis.

Table 7. The general scheme to support Phase 1.

Solid Matrix		**Analytical Method**	**Radionuclides**
Raw material	Raw material of natural origin	Gamma spectrometry	^{40}K, series of ^{238}U and ^{232}Th
	Raw material from industrial process	Gamma spectrometry	^{40}K, chain segments of ^{238}U and ^{232}Th
Residue	Dried Residue 1 (e.g., from refractory industry)	Gamma spectrometry Alpha spectrometry	^{40}K, chain segments of ^{238}U, ^{232}Th and ^{210}Po
	Dried Residue 2 (e.g., from cement production)	Gamma spectrometry	^{40}K, chain segments of ^{238}U and ^{232}Th
	Wet residue (e.g., oil sludge)	Gamma spectrometry	^{40}K, chain segments of ^{238}U and ^{232}Th

4.2.2. Phase 2

If the radiological content of radionuclides in solid matrices exceeds ELs in terms of activity concentration, the Phase 2 starts. Indeed, in this situation it is necessary to proceed with the assessment of doses to workers and members of the public to be compared with the relevant ELs in terms of effective dose. The four steps of Phase 2 are shown in Figure 3.

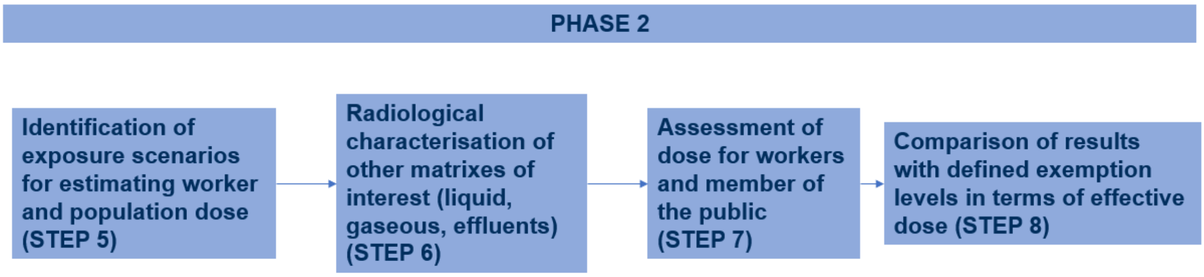

Figure 3. Steps of Phase 2 of the general methodology.

The assessment of dose for members of the public requires to consider many different exposure scenarios and pathways, including those involving liquid and gaseous effluents (steps 5); it is worth noting that, for workers, the selection of the most significant exposure scenarios takes into account the entire industrial process. Therefore, depending on the industrial process, it might be necessary to extend identification and radiological characterization of additional matrices of interest (steps 6).

In step 7, the dose assessment for workers and members of the public is carried out, taking into account all information, data and measurement results gathered in the previous steps. Finally, in step 8, the estimated doses are compared with ELs.

Table 8 presents the general categories of the exposure scenarios relevant to workers and members of the public; this is a useful tool for the application of Phase 2, likewise Table 7 is a supporting tool for the application of Phase 1.

Table 8. The general scheme to support Phase 2.

	Matrix	**Exposure Scenarios** **Workers**	**Public**
Industrial sector	Raw materials	Storage–Large quantity; transport; management	Transport
	Residues	Management (collection, loading, unloading, etc.)	Disposal (landfill, recycle, reuse, etc.); transport
	Final products	Finishing processes; packaging; transport	-
	Liquid effluents	-	Release of wastewater: exposure of members of the public to liquid effluents
	Gaseous effluents	-	Release of gaseous effluents from chimneys: exposure of members of the public living near the industrial plant

Specifically, Table 8 shows, for a generic industrial sector/practice involving NORM, the main categories of matrix of interest involved, and, for each of them, the possible significant exposure scenarios for workers and members of the public. It is worth noting that, limited to worker exposure, final products could be considered as matrix of concern since the finishing processes, or packaging or transport can be significant exposure scenarios.

In order to estimate the effective dose for workers, for each exposure scenario present in Table 8, Table 9 provide a detailed information to be gathered about matrices and the most relevant exposure pathways.

As regards the dose assessment for members of the public, the Table 10 plays the role covered by the Table 9 for workers. It is important to note that for this category of exposed individual effluents can give a significant contribution to dose, contrary to what happens for workers.

Table 9. Exposure scenario for dose assessment of workers.

Specific Exposure Scenarios	Matrix	Exposure Pathways
Exposure to heap, Transport, etc.	Raw material of natural origin	External exposure, inhalation, radon exposure
	Raw material from industrial process	External exposure, inhalation, radon exposure
Management of residues (Collection, loading, unloading, etc.) Transport	Dried Residue 1	External exposure, inhalation, radon exposure
	Dried Residue 2	External exposure, inhalation, radon exposure
	Wet residue (sludge)	External exposure, radon exposure
Finishing processes Transport	Final product	External exposure, radon exposure

Table 10. Exposure scenario for dose assessment of members of the public.

Specific Exposure Scenarios	Matrix	Exposure Pathways
Transport	Raw materials	External exposure, inhalation, radon exposure
Release from chimney	Gaseous effluent	External exposure, inhalation, secondary ingestion *
Release to water body or sewage system	Liquid effluent	External exposure, inhalation, secondary ingestion *
Exposure to residue with activity conc.> CLs	Sludge, grinding waste, dust from abatement plant, etc.	External exposure, inhalation, radon exposure

(*) Secondary ingestion of vegetables grown in the garden/farm

5. Conclusions and Perspectives

By adopting the graded approach, already recommended by ICRP publication 103, and endorsed by the EU-BSS and by the Italian legislation, a general methodology for characterizing the exposure scenarios relevant to NORM involving industries was developed. This methodology, made up of two phases each of which involves four consecutive steps, consists in a general approach which can be customized to fit a given industrial sector and/or scenario. In this paper some tables are presented as operational tools of the general methodology, in order to facilitate and coordinate an effective series of actions finalized to exempt or not a NORM involving industry. The application of the general methodology to some plants of different industrial sectors has been allowing the verification of its completeness and effectiveness. Results of these specific applications will be published in dedicated papers.

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